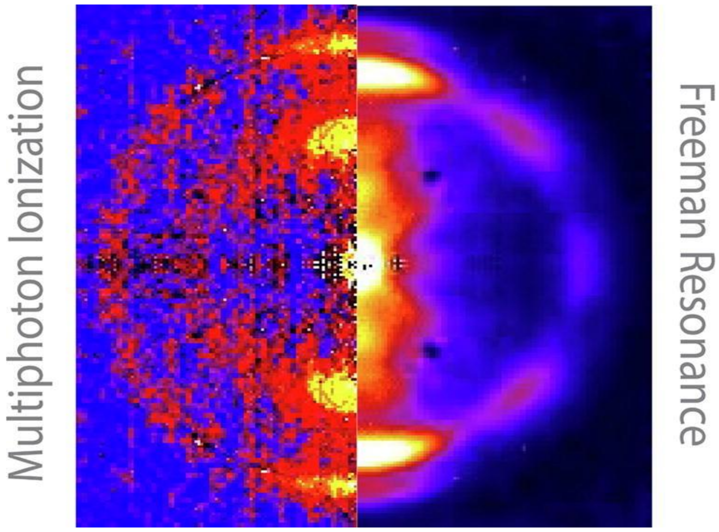
Differentiating Freeman Resonance and Strong Field Tunnelling Ionization

1. Overview of Freeman Resonance

Freeman resonance is a phenomenon observed in ultrafast laser physics, where a Rydberg state is AC-Stark shifted into multiphoton resonance with the atomic ground state. It is characterized by a strong peak in the electron kinetic energy spectrum, typically cantered around 0.7 eV. This resonance has a much larger ionization probability compared to non-resonant ionization processes. The key features of Freeman resonances include:

- **Kinetic Energy Consistency**: The kinetic energy of the electrons associated with the Freeman resonance remains roughly constant for laser intensities above a certain threshold.
- **AC-Stark Shift:** The primary effect is the AC-Stark shift, which shifts a Rydberg state into resonance with the atomic ground state.
- **Resonance Enhancement:** This enhancement is due to the bound-bound multiphoton transition.
- **Field Strength Independence**: The positions of Freeman resonances are almost independent of the field strength but are located below the ionization continuum due to the AC Stark shift.



- Experimental Observations

- **Freeman Resonance:** Experimental studies of Freeman resonance have observed specific kinetic energy peaks in photoelectron spectra. Examples include the femtosecond ionization of Methyl Iodide **(IP=9.5 eV)**, where a dominant resonance was observed at 800 nm and peak intensities of **≈ (1-6) × 10^13 W/cm^2 [Chem. Phys. Lett. 759,137984 (2020)]**.

- **Multiphoton Ionization:** MPI experiments have shown varied results depending on the target atoms and laser configurations. Experiments have demonstrated that MPI can produce electrons with energies corresponding to the absorption of multiple photons, leading to doubly charged ions and other high-energy states.

2. Overview of Strong Field Tunnelling Ionization

Tunnel ionization is a quantum mechanical process where an electron tunnels through the potential barrier of an atom or molecule due to the influence of an intense electric field. This phenomenon is often observed when interacting with near-infrared strong laser pulses. The essential characteristics of tunnelling ionization include:

- **Potential Barrier Distortion**: In an intense electric field, the potential barrier is drastically distorted, allowing electrons to tunnel through.
- **Keldysh Parameter**: The Keldysh parameter (γ) is utilized to distinguish between tunnelling ionization and multiphoton ionization (MPI), with $\gamma < 1$ indicating tunneling ionization and $\gamma > 1$ indicating MPI.
- **Quantum Mechanical Tunneling**: Unlike classical ionization where the electron needs enough energy to overcome the potential barrier, tunneling ionization allows the electron to pass through the barrier due to its wave-like properties.

Comparative Analysis

Ionization Mechanism

- **Freeman Resonance**: Enhances ionization through transient excitation of Stark-shifted bound states into multiphoton resonance with the ground state. The ionization probability significantly increases in this resonance condition.
- **Tunneling Ionization**: Occurs when electrons escape the potential barrier under the influence of a strong electric field due to quantum tunneling effects.

Sensitivity to Laser Intensity

- **Freeman Resonance:** The position of Freeman resonances is rather insensitive to the laser intensity and can persist despite variations in field strength.
- **Tunneling Ionization**: Highly sensitive to the intensity of the electric field. The tunneling probability increases exponentially as the field strength increases.

Spectral Features

- **Freeman Resonance**: Characterized by a strong peak in the kinetic energy spectrum, often around 0.7 eV, which remains constant above certain intensities.

- **Tunneling Ionization**: The electron momentum distributions and energy spectra are more broadly distributed and depend on the interaction of the electron with the field and the atomic potential.

Theoretical Framework

- **Freeman Resonance**: Often explained through the modification of Rydberg states by the AC Stark effect, leading to resonant multiphoton excitation.
- **Tunneling Ionization**: Theoretical models such as the **ADK (Ammosov-Delone-Krainov)** theory predict ionization rates based on the suppression of the potential barrier.

Key Distinctions

1. **Mechanism Dependence**:
    - **Freeman Resonance**: Dependent on multiphoton resonance and AC-Stark shift phenomena.
    - **Tunneling Ionization**: Governed by the quantum mechanical tunneling process influenced by the electric field intensity.
2. **Field Intensity Sensitivity**:
    - **Freeman Resonance**: Less sensitive to changes in laser intensity.
    - **Tunneling Ionization**: Highly sensitive to the field intensity, with ionization rates increasing sharply with higher field strengths.
3. **Wavelength Sensitivity:**
    - **Freeman Resonance**: highly sensitive to changes in laser wavelength or photon energy as resonance energy shift changes the ionization rate.
    - **Tunneling Ionization**: Less sensitive to the field intensity.
4. **Kinetic Energy Characterization**:
    - **Freeman Resonance**: Exhibits a distinct peak at a specific kinetic energy.
    - **Tunneling Ionization**: Produces a broader spectrum influenced by the interaction dynamics during ionization.

By understanding these key distinctions we can accurately differentiate between Freeman resonance and strong field ionization by tunneling ionization. If the ionization yield is same for different laser wavelength then, we can confirm that it is a strong field ionization by tunnelling, if changes with wavelength/energy then it is a resonance.